PROJECT INDALO

WORK PERFORMED DURING 2001

SUMMARY

On September 15, 1997 a new agreement between the U.S. Department of Energy (DOE) and the Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT) was signed to establish a framework for scientific and technological cooperation by the Parties in radiological studies resulting from the accidental release of fissionable materials that occurred in Palomares, Spain, on January 17, 1966.

As a consequence of the confluence of a set of specific circumstances, a change in the management of the Palomares situation is being negotiated. The following reasons can be argued:

- A Pu-239+240 inventory higher than the previously estimated in the zone of maximum residual contamination (zone 2-0).
- Existence, in this zone, of a burial of contaminated wastes from inmediate cleaning operations after the accident, which is still not located. There is no information about further actions of decontamination or removal of these wastes.
- Given the potential for new agricultural practices and the available water irrigation resources in the zone, the owners of the land have expressed their intention to cultivate the zone. This would lead to a more extensive and intensive soil use.
- Growing activity of Am-241, due to the passage of 36 years since the accident took place.
- Recent data which demonstrate the existence of contaminant hot particles, involving potential changes in the behavior of the residual contamination.

The new management plan involves a process of conveyance of ownership (from the current owners to an administrative body) of the lands where the higher residual contamination levels from the 1966 accident remain. This process is essential in order to assure continuation of further research activities in the more affected zones. The most suitable way to convey the ownership is being discussed among representatives of the national regulatory body Consejo de Seguridad Nuclear, CIEMAT, ENRESA (National Agency for the Management of Radioactive Wastes) and the municipality of Palomares. Then, a (no longer than 10 years) research plan will be carried out. In addition to scientific objectives, leading to improve the knowledge on environmental and metabolic behavior of Pu and Am, the final goal of the research plan is to determine the most suitable options for environmental restoration. Obviously, this should be based on risk assessments under present conditions and other potential future use scenarios.

In parallel, during 2001, environmental and personal radiological monitoring activities have been continued. Also, according to the work proposal for the year 2001 and following the recommendations made by the expert panel in its 1998 report, other studies mainly related to soils have been carried out. Soil studies are focussed in Zone 2.0, where the highest levels of contaminants exist, but also in Zone 3, where more recently some metallic fragments, probably from the bomb 3 materials or components have appeared. Activities dealing with the extension and refinement of risk assessment have been deferred to be included in the research plan cited above.

The activities performed in 2001 are detailed in the two semi-annual reports sent to the Spanish national regulatory body, Consejo de Seguridad Nuclear (CSN). These two reports are attached (1), (2).

Soils

Isotopic composition of the bombs fuel

Recently, an estimation of the isotopic composition of the bomb fuel was made, from contaminated soil samples from zone 2-0. Given that the zone 2-0 was initially contaminated with the plume of the bomb 2, and was also used as operative zone to manage the contamination originally removed from the zone 3, the estimation would correspond to a mixture of both bombs 2 and 3. In addition some evidence suggests that the two bombs could be different in composition. Therefore, a specific estimation of the isotopic composition of each of the bombs should be made. To do it, a selective soil sampling, based on the location of points with high radiometry, has been planned and carried out in the zone 3 (where only contamination of bomb 3 exists). Soil samples from zone 2-0 have been specially selected from parts of this zone where only contamination from bomb 2 may be guaranteed (unaltered hill landscape). Then, a removal of radioactive particles from the earlier mentioned different samples will be made in order to determine isotopic ratios between Plutonium isotopes and between Plutonium and Americium concentration.

A small number of samples have been analyzed so far, two of them coming from zone 2-0 and the rest (three) coming from soil adhered to one of the fragments found in zone 3. This study is being continued in order to obtain a statistically significant number of results, which will be reported when completed.

Dispersion of the contamination outside the originally marked zero line

Since the time of the accident, some physical processes, such as wind, traffic, etc. could have conducted to a dispersion of the contamination from the initially contaminated areas to other initially clean areas. The objective of this activity is to monitor and determine the present existing level of contamination beyond the initially considered zero line of contamination.

Fifteen surface soil samples collected, covering a 50m x 50m square surface, from the so-called zone 5-3B (initially considered as contamination free area) in 1999 were analysed for Pu 239+240. The results are shown in table 1. Only 4 of the 15 samples showed positive values of Pu, above the lower detection limit (LLD),

with a maximum value of 5.5 Bq/kg. In spite of these low values, the zone can not be considered as totally free of contamination.

Table 1. $^{239+240}$ Pu in surface soil samples (1999 sampling, zone 5-3B).

Sample	$Pu^{239+240}$ Activity Bq/kg
1	1,36 ± 0,33
2	1,25 ± 0,28
3	5,50 ± 0,77
4	≤ 0,44
5	≤ 0,80
6	≤ 1,00
7	≤ 0,98
8	≤ 1,33
9	≤ 0,38
10	≤ 0,35
11	≤ 0,35
12	≤ 0,35
13	1,07 ± 0,20
14	≤ 0,35
15	≤ 0,35

Also, two samples from outside the area 2 but close to the zone 2-0 and potentially affected by its runoff, were collected in November 2001 and then analyzed, showing values of Pu of about 27 kBq/kg and 72 kBq/kg. The confirmation of the magnitude and extension of this contamination and its cause will require further effort.

Inventory of Am-241

A review of the data obtained from a 1986 soil sampling (45cm depth), corresponding to 5.5 hectares in the zone 2-0, was made. The values of Am concentration in five depth intervals is shown in table 2. The inventory estimate has been made following the methodology used for Pu inventory estimate at the same area. An estimate of about 11.5 Ci (more than 4 E11 Bq) has been obtained for the mentioned date. A detailed description of the methodology is given in the document attached to this report (3), where the location of the 37 sampling points in relation to its topographic coordinates can also be seen.

Table 2. Am-241 (kBq kg^{-1}) at different depth intervals in zone 2-0

Point	Position	Depth (cm)				
		0 - 5	5 -15	15 - 25	25 - 35	35 - 45
1	270-117	167	164	314	9,09	35,7
2	300-110	7,62	5,32	6,39	4,28	2,71
3	287-100	232	103	59,9	37,3	120
4	330-100	0,97	0,77	0,26	1,44	0,13
5	285-90	21,6	19,2	16,2	4,52	1,5
6	271-90	3,02	< 0,009	< 0,01	< 0,01	< 0,009
7	310-90	1,85	1,73	4,76	1,66	0,47
8	297-70	30,41	49,62	32,74	1,55	2,49
9	330-70	0,18	0,11	0,18	0,05	0,02
10	290-50	14,6	9,68	1,56	2,79	1,03
11	285-30	7,32	3,48	0,69	1,06	0,38
12	290-123	13,27	20,73	11	12,9	1,16
13	320-123	0,7	1,57	1,13	0,19	0,4
14	310-145	47,9	5,14	3,54	0,4	0,64
15	250-145	62,4	43,9	51,9	36,3	1,24
16	260-145	7,6	3,21	2,25	0,17	0,44
17	280-160	6,91	0,94	0,17	0,5	< 0,008
18	300-160	51,23	172	18,9	3,86	0,03
19	310-160	49	24,7	12,3	0,24	< 0,014
20	320-160	1,65	0,91	0,71	0,02	< 0,01
21	310-180	2,51	2,24	23,6	0,269	0,06
22	300-180	31,8	6,36	5,46	3,48	8,93
23	280-180	6,43	8,94	3,45	< 0,007	< 0,01
24	285-200	2,73	0,24	0,04	< 0,006	0,07
25	300-200	1,18	1,61	1,04	0,14	0,005
26	320-200	6,62	3,81	3,01	0,7	< 0,006
27	310-225	15,5	29,3	30,6	12	0,76
28	300-225	16,8	10,1	3,97	3,58	0,11
29	290-225	5,33	13,3	0,81	4,29	1,56
30	325-225	6,31	7,5	24,6	9,38	0,19
31	335-225	54,64	1,66	< 0,008	< 0,01	< 0,014
32	300-240	5,07	4,71	10,4	4,04	0,29
33	360-240	6,61	< 0,009	< 0,011	< 0,006	< 0,01
34	290-270	3,77	3,81	10	8,98	2,2
35	330-270	29,1	1,82	< 0,011	< 0,008	< 0,011
36	350-300	58,7	< 0,009	< 0,01	< 0,009	< 0,008
37	325-340	41,6	0,23	< 0,007	< 0,005	< 0,01

Activity size distribution

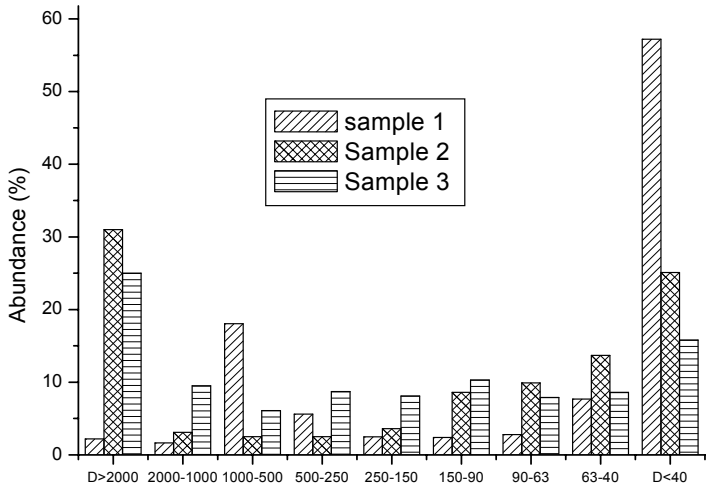
The high level of hetereogeneity in the distribution of the contamination in soils, previously confirmed, could change with time. This is why a new analysis of the activity distribution by size particles has been initiated during 2001. This study should provide the evolution over time of the active particles due to physical or chemical mechanisms. The potential fragmentation of these particles would lead

to make them more easily inhaled due to resuspension processes, increasing the radiological risk. Three soil samples from zone 2-0 have been selected and its granulometric analysis has been made (see table 3 and figure1). Activity concentration in each of the size fractions is being determined.

Table 3. Granulometric fractions and weight percentages

Granulometric fraction	weight percentage (sample 1)	weight percentage (sample 2)	weight percentage (sample 3)
$\phi > 2000\mu$	2,18	31,0	25,0
$1000\mu < \phi < 2000\mu$	1,64	3,1	9,5
$500\mu < \phi < 1000\mu$	18,04	2,5	6,1
$250\mu < \phi < 500\mu$	5,6	2,5	8,7
$150\mu < \phi < 250\mu$	2,48	3,6	8,1
$90\mu < \phi < 150\mu$	2,38	8,6	10,3
$63\mu < \phi < 90\mu$	2,79	9,9	7,9
$40\mu < \phi < 63\mu$	7,68	13,7	8,6
$\phi < 40\mu$	57,21	25,1	15,8

Figure 1. Weight abundance in the granulometric fractions of the samples



Research on Pu hot particles in soils

There is still a lack of knowledge to fully understand the processes of hot particle formation, spatial density and local distribution in soil, mechanic transport (water,

wind), migration by dissolution processes and composition. This knowledge will enable to predict their environmental and metabolic behavior, the risk involved and the assessment of health hazards.

Further to the findings of hot particles and metallic fragments reported last year, a new search was carried out in zone 3 during November 2001. The search was performed by a foot radiometric survey covering a surface of this zone located in the opposite direction to the original contamination plume of the bomb 3. Seven new highly radioactive fragments have been found and collected. These fragments will be treated in order to separate the soil dust and radioactive particles easily removed.

The field methodology applied in order to locate samples containing hot particles is based on previous radiometric survey data, obtained with a mobile unit, which enables us to know the average exposition rate of the Palomares area and the corresponding standard deviation. In a first step, radioactive particles are searched in places where the exposition rate is higher than the average value. The criterion to collect a sample is to find spots with exposition rate three times higher than the average value.

The following equipment has been used for the field methodology*:*

- Total counting scintilometer SPP-2 digitized and mounted in a mobile unit. Data are collected every five seconds and sent to a portable PC jointly with the geographical position.

- Total counting scintilometer SPP-2 for foot survey.

- Dose-ratemeter model ES-3 for correlation between exposure and dose rates

- Gamma portable spectrometer, EXPLORANIUM GR-130, for nuclides identification at the field

A methodology to isolate radioactive particles in the laboratory has also been developed and applied. Confirmation of the presence or absence of hot particles into the different matrices (soil, vegetation and animal ashes, etc.) is previously made by autorradiography of the sample. Once the presence of hot particles is confirmed in a soil sample, two separation processes are combined in order to concentrate their density: particle size fractionation and binary separation. Each soil fraction and sub-fraction is then measured by a gamma-Ge detector to investigate differences in americium activity. The procedure of cutting and counting is repeated until the desired small fraction size containing the hot particle is reached. Finally, it is possible to obtain single particles in which the 99% of the activity is contained. Then, they are fixed onto a carbon tape for further studies.

There is a need for reliable techniques leading to determine morphology, composition and structure of the isolated radioactive particles. Different techniques are being applied:

- Optical microscopy, used to see the colour, brightness and shape.

- Electronic microscopy, allowing to confirm the presence or absence of uranium, plutonium and americium, as well as the morphology of the particles.

- X-ray emission techniques (i.e. μ-X-ray fluorescence -μ-XRF-, μ-particle-induced X-ray emission -μ-PIXE- and electron probe microanalysis –EPMA-): essential for the knowledge of the elemental composition, physico-chemical structure, including chemical bonding, oxidation states, structure of crystalline and amorphous phases and interfaces of the hot particles.

Research based on the above techniques is currently in progress. The inherent complexity of this research and the high specific activity involved requires a big care in handling the particles. Time spent and effort are therefore higher than initially expected.

Other soil activities

A systematic sampling of zone 3 was carried out during November 2001 following a square network and covering a total of 24 points from a surface of 2,465 m^2, located in the direction opposite to the original contamination plume of the bomb 3. The sampling also covered a small part of the zone 5, just close to zone 3. The objective of this sampling is to better characterize this part of the zone, which was never studied before 2000. In addition, the sampling should provide suitable samples to be used in the isotopic composition study previously mentioned. Analysis of the collected samples is currently in progress.

Simultaneously to this sampling a foot-radiometric survey was performed covering the total surface mentioned earlier to obtain further information about the existence of additional hot particles and fragments, from the bomb 3, to those found in a previous radiometric survey (year 2000).

Also, a sampling of the so called zone 6, never studied so far has been started in November 2001, collecting 7 surface soil samples, in order to determine the potential contamination level in this zone. In addition a foot-radiometric survey was performed in this zone 6, covering a surface of about 24,000 m^2. This survey detected Am-241. Based on the results of the 7 samples collected, the quantification and variabilty of the contamination in this zone will be estimated.

Air

In relation to the air monitoring, the sampling of air particles at stations 2-1, 2-2 and P have continued, changing the filter on a weekly basis. The air volume collected is about 10,000 m^3 per sample in average. The air sampler in station 2-0 continues to be out of service from December 1999 due to break of the air aspiration pump. The replacement of the air pump by a new one has not been successful because the new device did not meet the technical specifications required. The air pump was then sent back to the supplier in USA and we are waiting for the appropriate new one. In addition some difficulties appeared with the owner of the parcel where the station 2-0 is located in order to operate the station, although it seems to be now resolved. The new station 2-1, which was

placed in a more adequate Zone 2-1 site (April 2000), failed on 15 May 2001 and restarted on 26 October 2001. Station P has been working during all the year 2001, with the exception of only a week (12 to 20 January) due to electrical supply problems. Station 2-2 worked without interruption. In total 130 air samples were collected during 2001.

Samples from 2000 have been analyzed and measured for Pu-239+240. Average monthly values of Pu ranged, during 2000, from 0.5 to 21.6 μBq m^{-3} in station 2-2, from 2.0 to 42.2 μBq m^{-3} in station 2-1 and from 0.2 to 6.1 μBq m^{-3} in station P. In general, these values are in the range of the historical data series. Also, in agreement with the general trend, seasonal variability is higher than the annual one, as it could be expected considering seasonal weather changes and the timing of agricultural practices. The results pertaining to the year 2000 are shown in the tables 4, 5 and 6.

Table 4. Pu-239+240 in Station P. Year 2000

Sampling Period	Pu-239+240 Activity μBq/m^3
01-01-2000 to 11-03-2000	—
11-03-2000 to 31-03-2000	0,22 ± 0,08
31-03-2000 to 28-04-2000	0,44 ± 0,11
28-04-2000 to 02-06-2000	0,29 ± 0,08
02-06-2000 to 30-06-2000	1,40 ± 0,40
30-06-2000 to 28-07-2000	0,7 ± 0,18
28-07-2000 to 01-09-2000	6,10 ± 1,40
01-09-2000 to 29-09-2000	0,90 ± 0,20
29-09-2000 to 04-11-2000	0,48 ± 0,12
04-11-2000 to 01-12-2000	1,00 ± 0,20
01-12-2000 to 29-12-2000	1,10 ± 0,30

Table 5. Pu-239+240 in Station 2-1. Year 2000

Sampling Period	Pu-239+240 Activity $\mu Bq/m^3$
01-01-2000 to 14-04-2000	—
14-04-2000 to 05-05-2000	22,6± 2,3
05-05-2000 to 02-06-2000	2,0± 0,5
02-06-2000 to 30-06-2000	17,7+ 4,1
30-06-2000 to 04-08-2000	3,9± 0,5
04-08-2000 to 01-09-2000	2,0± 0,5
01-09-2000 to 29-09-2000	2,9± 0,7
29-09-2000 to 04-11-2000	4,9± 1,1
04-11-2000 to 01-12-2000	42
01-12-2000 to 29-12-2000	4,8± 1,1

Table 6. Pu-239+240 in Station 2-2. Year 2000

Periodo de muestreo	Actividad de Pu^{239} $\mu Bq/m^3$
31-12-1999 to 28-01-2000	0,47± 0,13
28-01-2000 to 03-03-2000	1,20 ± 0,30
03-03-2000 to 31-03-2000	2,10± 0,50
31-03-2000 to 28-04-2000	4,80± 1,10
28-04-2000 to 02-06-2000	3,40± 0,80
02-06-2000 to 30-06-2000	21,60± 5,00
30-06-2000 to 04-08-2000	3,87 ± 0,52
04-08-2000 to 01-09-2000	5,10 ± 0,58
01-09-2000 to 29-09-2000	8,69 ± 1,01

29-09-2000 to 04-11-2000	5,27 ± 1,02
04-11-2000 to 01-12-2000	2,05 ± 0,32
01-12-2000 to 29-12-2000	2,86 ± 0,39

Vegetables

For vegetables, 18 samples of different crops have been collected in 2001. These samples are further divided in different parts (leaves, edible, etc.) and pretreated for analyzing separately in order to determine the type of contamination (external or internal) and its distribution.

Samples of vegetables collected during 2000 have been processed for Pu, involving 19 analyses. These samples were collected in Zones 2, 3 and 5 (concretely 5-3B). With the exception of the leaves of watermelon samples, all the values were ≤ 1 Bq kg^{-1}; in general, values were higher for leaves than for fruits. Also 4 Pu analyses of samples collected in 2001 (1 corresponding to wheat from zone 3 and 3 corresponding to wild vegetation from zone 5) have been performed, giving values higher than 1 Bq/kg (3.6, 17.7, 11.2 and 12.4 Bq/kg respectively). These values in samples from zone 5 suggest these to be due to resuspension of particles from other more contaminated areas.

Concerning Am-241, due to the high LLD obtained by low energy gamma spectrometry, the analysis by radiochemistry separation followed by alpha spectrometry measurements was decided. Only two samples were analysed, one from zone 2-0 (watermelon) and another one from zone 3 (tomato), giving results below the LLD, in the order of a few mBq/kg.

The small number of vegetables processed during 2001, smaller than in preceding years has been due to the installation of new lab boxes in the related laboratory.

The proposed historical review analysis has been postponed due to other work priorities.

Other products

Concerning milk samples, 10 samples were collected, analyzed and measured for Pu, 5 of them corresponding to cow milk and the other 5 corresponding to goat milk. The results are shown in table 7 and present values lower than LLD or in the order of a few mBq/l. Also analyses of Am (radiochemistry followed by alpha spectrometry) have been performed in two of the mentioned samples, giving values lower than LLD.

Two samples of snails from zone 2-0 were also collected during 2001

Table 7. Pu239+240 in milk samples

Type of sample	Sampling date	Density g/cc	Volume analysed ml	$Pu^{239+240}$ Activity mBq/l
Cow milk-1	15-9-01	1,03	294	0,97 ± 0,35
Goat milk-1	15-9-01	1,03	236	4,78 ± 0,99
Cow milk-2	24-9-01	1,14	245	1,87 ± 0,49
Goat milk-2	24-9-01	1,05	218	1,30 ± 0,43
Cow milk-3	1-10-01	1,06	263	≤ 0,50
Goat milk-3	1-10-01	1,02	237	≤ 0,50
Cow milk-4	8-10-01	1,03	280	≤ 0,16
Goat milk-4	8-10-01	1,02	256	≤ 0,30
Cow milk-5	15-10-01	1,07	263	≤ 0,68
Goat milk-5	15-10-01	1,01	241	1,81 ± 0,41

Urine

During 2001, 147 people from Palomares were transported to CIEMAT headquarters in Madrid for medical examinations and sampling of 24-hour urine collections and further bioassay analyses for internal dosimetric assessments. All those whose urine samples tested positive values, are requested to return to Madrid during the following year to confirm the existence and magnitude of their internal contamination. However, since the examinations are performed only on those who agree to participate, not all those requested for follow-up examinations are re-evaluated during the following year. Some individuals choose not to participate for different (personal) reasons.

In relation to Pu, the measurements of the 147 people examined during 2001 were completed. Only one person, corresponding to a child 14 years old, resulted in a positive value (very close to the lower detection activity of the analytical method) of 0.38 mBq in urine of 24 hours. It is the first time that this person is controlled

and therefore he will be requested to return the next year for a new control. Based on the results of this future analysis, the related dosimetric assessment will be performed.

The percentage of positive values for the year 2001 (0.68 %) is lower than the percentage for the period 1967-2000 which is 4.03 % (143 positive values in a total of 3,546 measurements). The distribution by ranges of these results, for the total period, approach to a log-normal function; considering the median of the distribution as the most representative parameter for the more frequent committed (50 years) effective dose in the group of people having dose estimation, a value lower than 2mSv per year is obtained. The results obtained in 2001 do not modify the conclusions from the expert panel in its 1998 report.

Concerning Am, the urine analyses of the 147 people examined during 2001 were also completed. 146 samples showed values below the detection limit of 0.37 mBq in urine of 24 hours. A 29 year-old man (agricultural worker) presented a value of Am in urine of 0.56 mBq. This man had been analyzed before on 8 occasions, this being the first time that contamination was detected in his urine. This person will be also requested to return the next year for a new control, in order to make their dosimetric assessment in the case of a new positive result. During 2001, no measurements in the whole body counter were made.

The individual results of bioassay for each of the person who are annually examined, were registered and sent to each of them, jointly with the result of their medical examination. These individual data are confidential. No significant findings related to radiation exposure were reported concerning the medical examinations (147 people) performed during 2001.

ATTACHED DOCUMENTS

(1) Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Primer Semestre del Año 2001). CIEMAT/DIAE/PPRI/51100/02-2001).

(2) Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Segundo Semestre del Año 2001). CIEMAT/DIAE/PPRI/51100/02-2002).

(3) A. Espinosa, A. Aragón, B. de la Cruz. "Estimación del contenido de Americio existente en el año 1987 en una zona de Palomares contaminada en 1966 por material de Plutonio grado bomba". Colección Informes Técnicos Ciemat nº 975, Octubre 2001.

(4) A. Espinosa, A. Aragón, B. de la Cruz, J. Gutiérrez. "Estudio de la contaminación por Plutonio y Americio en un área agrícola. Impacto radiológico ocasionado por consumo de vegetales contaminados". Fifth Congress on Radiation Protection and Safety - IRPA Regional Congress. Recife – Brasil. April 29 - May 4, 2001.

(5) A. Aragón, A. Espinosa, J. A. Fernández. "La presencia de Americio en el suelo de Palomares después de 33 años del accidente, y su influencia radiológica". Fifth Congress on Radiation Protection and Safety - IRPA Regional Congress. Recife – Brasil. April 29 - May 4, 2001.